

TERRESTRIAL RADIOACTIVITY IN HUNGARIAN ADOBE BUILDING MATERIAL AND DWELLINGS WITH A FOCUS ON THORON (^{220}Rn)

by

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Résumé

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1. INTRODUCTION AND OBJECTIVES

Radon (^{222}Rn) and thoron (^{220}Rn) from the terrestrial ^{238}U and ^{232}Th decay chains are responsible for about the half of the total annual effective dose from natural sources (UNSCEAR 2000). In the past, exposure to thoron was often ignored due to its very short half-life. It is already known that it can significantly contribute to the radiation dose in certain environments (UNSCEAR 2006). Several studies (e.g. Yonehara et al. 2005) show elevated radon and thoron activity concentrations in dwellings built of soil and mud which are very similar to Hungarian adobe dwellings. The main objective of this research was to evaluate the terrestrial radiation risk from the adobe building material used in Hungary. I focus on their resulted external and internal radiation exposures and it is also aimed to help later studies to find high risk localities and periods. I also provide solutions for problems occurred in the radon and thoron emanation determination methods, which is the measure of radon and thoron recoiled or diffused to the pores of the building material.

2. REFERENCE LEVELS

To limit external exposure in dwellings, different building material hazard indices are used. The ones to best consider are the radium equivalent (Ra_{eq}) and activity concentration (I) indices with threshold values of 370 Bq kg^{-1} and 1, respectively, which lead to about 1 mSv y^{-1} effective dose (EC 1999). Regarding internal exposure, the World Health Organization (WHO 2009) declares that the chosen reference level should not exceed 300 Bq m^{-3} radon activity concentration. This corresponds to about 10 mSv y^{-1} order of magnitude effective dose (ICRP 2009). Reference value does not exist for thoron gas itself. I decided to compare these results to the same activity concentration of 300 Bq m^{-3} .

3. STUDIED AREAS

Three distinct areas in Hungary were selected to study. These areas are Békés County (SE-Hungary), which the most typical area of average Hungarian adobe dwellings, the E-Mecsek Mts. (S-Hungary) which is well-known for its granite bedrock and the Sajó and Hernád Rivers Valleys (NE-Hungary). Nineteen settlements were selected to collect 46 adobe building material samples in total. The in-situ measurement campaigns were performed only at Békés County in its 53 adobe dwellings.

4. METHODS

Laboratory methods applied on 46 adobe building material samples from Békés County, E-Mecsek Mts. and Sajó and Hernád Rivers Valleys:

- radon emanation determination by RAD7 detector with the comparison of “equilibrium” and “growth curve (ingrowth)” measurement protocols, sensitivity analysis to radon leakage from the experimental setup, thoron emanation determination by RAD7 detector in an improved data analysis method taking into account the sample geometry and the thoron attenuation in the sample holder,
- ^{226}Ra , ^{232}Th and ^{40}K activity concentration determination by γ -ray spectrometry and related radon and thoron emanation fraction, building material hazard index (Ra_{eq} and I) calculations and external effective dose estimation,
- grain size distribution determination by wet sieving and laser grain size analysis and related specific surface area estimation.

In-situ methods applied in 53 adobe dwellings at Békés County:

- indoor radon and thoron activity concentration determination at 10 cm distance of adobe walls by track etched detectors in four seasons and related inhalation (internal) dose estimation,
- equivalent γ dose rate determination by FH 40 G-L10 portable device.

Advanced statistical methods applied for data evaluation:

- test of differences between central tendencies of sample groups by Mann-Whitney (Wilcoxon) hypothesis tests,
- test the normal and lognormal distribution assumptions of data by Shapiro-Wilk hypothesis tests,
- correlation analysis by Pearson's linear correlation coefficients.

5. THESIS

1. I have tested a time saving – for large number of samples – radon emanation measurement method, the so-called equilibrium method regarding its sensitivity to the radon leakage from the experimental setup, which cannot be measured inside the method. I used another radon emanation measurement method, the growth curve (ingrowth) method to compare, which provides a value for the radon leakage rate in addition. The results show that for the appropriate usage of the less time consuming equilibrium method, a proven maximum $0.0025\text{--}0.003\text{ h}^{-1}$ radon leakage rate (α , about 30–40 % of the value of radon decay constant) has to be ensured by the design (careful sealing) of the experimental setup (Szabó et al. 2011).

2. I have described in an experiment a non-linear RAD7 detector thoron activity concentration response as a function of the thickness of a cylindrical sample. Thus, I have contributed to the final form of a model which is necessary in thoron emanation determination taking into account the sample geometry, the thoron decay in RAD7 detector and the resulted thoron activity concentration attenuation in the sample container. This model matches my experimental results, and provides an estimate for the thoron diffusion coefficient (D) in adobe building material, which is in the range of 1 to $3 \times 10^{-6}\text{ m}^2\text{ s}^{-1}$ (Csige, Szabó et al. 2013). I applied this value in the thoron emanation determination of other samples.

3. I have verified that the external radiation of adobe building material does not carry any radiation risk. The building material radiation hazard indices considered (radium equivalent index, Ra_{eq} and activity concentration index, I) for the 46 adobe samples – from Békés County, E-Mecsek Mts. and Sajó and Hernád Rivers Valleys – are far below their given threshold values (370 Bq kg^{-1} and 1, unit) and also lower than for similar types of building materials reported in other countries. Both estimated annual external effective doses and in-situ measured γ dose rates confirm that the building material excess in adobe dwellings is in all cases below the accepted criterion of 1 mSv y^{-1} (Szabó et al. 2013).
4. I have concluded that the inhaled radon, thoron and their progenies present an important internal radiation risk in adobe dwellings. I have measured annual indoor radon and thoron activity concentrations at 10 cm distance from adobe walls with medians of 188 and 232 Bq m^{-3} , respectively. Accepting lognormal distribution I have demonstrated that 14-17 % of the adobe dwellings at Békés County have radon activity concentration higher than the WHO reference level of 300 Bq m^{-3} . For comparison, accepting normal distribution, 29-32 % of them will have higher thoron activity concentration than the same value. I have estimated the radon inhalation doses to exceed 10 mSv y^{-1} in 7 % of the 53 studied adobe dwellings and additionally thoron contributes with an elevated estimated average of 30 % in the total inhalation dose (Szabó et al. in press).
5. I have stated that the adobe building material acts as generally important radon and thoron source for the reason of its high radon and thoron emanation fractions. I have measured high radon and thoron emanation fractions (27 and 18 %, respectively) but comparably low ^{226}Ra and ^{232}Th activity concentrations (28 and 32 Bq kg^{-1} , respectively, Szabó et al. 2013) for the 46 adobe samples from Békés County, E-Mecsek Mts. and Sajó and Hernád Rivers Valleys.
6. I have pointed out that among determined parameters, the ^{226}Ra and ^{232}Th activity concentrations, radon emanation fraction and estimated specific

surface area show significant differences for adobe sample groups originated from different distinct areas of Békés County, E-Mecsek Mts. and Sajó and Hernád Rivers Valleys with different geological backgrounds. In this study, the ^{226}Ra and ^{232}Th activity concentrations in adobe building materials are elevated on the loess covered E-Mecsek Mts. with granite bedrock (Szabó et al. 2013). The radon emanation fractions are the highest at Sajó and Hernád Rivers Valleys. However, the specific surface area estimated from grain size distribution is significantly higher at Békés County.

7. I have observed that the in-situ measured radioactivity levels at Békés County, i.e. the annual indoor radon and thoron activity concentrations and the average γ dose rates have characteristic spatial distributions on the type of local Quaternary sedimentary formations. Among clay, loess and turf, the highest values are always observed and hence can be expected on clay formations (Szabó et al. in press). Comparing the age of the same formations, Pleistocene, Holocene, values slightly higher (<95 % confidence level) are detected on Holocene age formations.

8. I have described and explained different seasonal indoor activity concentration variations for radon than for thoron. Radon median displays a close to typical seasonal variation with high values in winter and autumn, lower values in spring and low values in summer. At the same time thoron median is steadily decreasing during the measurement period from winter to autumn. I have also presented different statistical distribution (lognormal, normal) variations for the two isotopes. Based on these results I have pointed out that the radon data follow the average temperature changes and are affected by the increased ventilation in summer and I observed that seasonal thoron data is moving together with the amount of precipitation (moisture content of adobe building material) through a one year measurement period (Szabó et al. in press).

SELECTED PUBLICATIONS

- Szabó, Zs.**, Jordan, G., Szabó, Cs., Horváth, Á., Holm, Ó., Kocsy, G., Csige, I., Szabó, P. & Homoki, Zs. (**in press**) Radon and thoron levels, their spatial and seasonal variations in adobe dwellings – a case-study at the Great Hungarian Plain. *Isotopes in Environmental and Health Studies*, DOI:10.1080/10256016.2014.862533
- Csige, I., **Szabó, Zs.** & Szabó, Cs. (**2013**) Experimental technique to measure thoron generation rate of building material samples using RAD7 detector. *Radiation Measurements* 59, 201–204.
- Szabó, Zs.**, Völgyesi, P., Nagy, H.É., Szabó, Cs., Kis, Z. & Csorba, O. (**2013**) Radioactivity of natural and artificial building materials – a comparative study. *Journal of Environmental Radioactivity* 118, 64–74.
- Szabó, Zs.**, Szabó, Cs. & Horváth, Á. (**2011**) Comparison of two ^{222}Rn mass exhalation rate measurement methods by study of Hungarian adobe building materials. Nordic Society for Radiation Protection Conference Proceedings, Reykjavik, <http://www.nsfs.org/NSFS-2011/documents/session-12/S12-O7.pdf>

REFERENCES

- EC (1999) Radiation Protection Report No. 112.,
- ICRP (2009) ICRP Ref 00/902/09.,
- UNSCEAR (2000) Annex B, 83–156. in: UNSCEAR Report.,
- UNSCEAR (2006) Annex E, 197–334. in: UNSCEAR Report.,
- WHO (2009) WHO Press, ISBN: 978 92 4 154767 3.,
- Yonehara et al. (2005) International Congress Series 1276, 58–61.